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SERIAL No. 12

(11) 4 Jan 1943

(14) NRL-1977

(12) 416.

(9)

NAVY DEPARTMENT

Progress Report on Liquid Thermal  
Diffusion Research.

(1)

NAVAL RESEARCH LABORATORY  
ANACOSTIA STATION  
WASHINGTON, D. C.

Number of Pages:

Text - 23

Plates - 11

Authorization:

BuShips Project Order 47/41 of 7/10/40

BuShips ltr C-NP14/L1-2(7-13-SS) of 7/16/40

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ABSTRACT

A study has been made of the liquid thermal diffusion method of separating uranium isotopes. In the course of the research 29 diffusion columns have been built and tested. At present a 14 unit pilot plant is undergoing tests. Separations between end fractions of 45.5% have been obtained by the use of two 48 foot columns connected in series. This series pair is capable of producing two fractions of  $UF_6$ , 1 kg of each, possessing an isotope separation factor of 1.2.

Work to date shows the apparatus to be unusually dependable and capable of long time trouble free operation. Separation of uranium isotopes on a large scale by the method is a practical possibility. The initial cost for a 1 kg 90%  $U_{235}$  per day plant is estimated at ~~\$25,000,000~~. The various other methods of isotope separation are discussed. Liquid thermal diffusion compares favorably with any of them.

An alternative to the 1 kg 90%  $U_{235}$  plant is presented. This involves the preparation of large amounts of 1.6%  $U_{235}$  and its use by the Chicago group.

Potentialities for improvement of separation factor and relaxation time have not been exhausted. Further research work is desirable before the design of a large plant is attempted.

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INTRODUCTION

(a) Authorization

1. This report is authorized by BuShips Project Order 47/41 of 7/10/40, and BuShips ltr C-NP14/L1-2(7-13-SS) of 7/16/40.

(b) Statement of Problem

2. In January 1939 a new vista was opened in science. For the first time the possibility of tapping atomic energy became very real. Experiments showed that a Uranium atom can be disintegrated by a neutron. In this process, three neutrons are given off together with 200,000,000 electron volts of energy. If one pound of Uranium could be thus disintegrated, a liberation of energy equivalent to that set free in the burning of 1370 tons of coal would result. Investigations were begun in many laboratories to accumulate more information about the process. It was found that only one of the isotopes of Uranium ( $U_{235}$ ) can be readily disintegrated in this way. Moreover,  $U_{238}$  which constitutes 99.3% of ordinary Uranium absorbs neutrons without being disintegrated, thus acting to prevent the formation of an explosive chain reaction. Means were proposed for decreasing this absorbing effect and indeed these give promise. However, in order to liberate energy using ordinary Uranium it is necessary to assemble a pile of Uranium and graphite weighing more than 100 tons in a special geometrical form. Experiments on such a pile are in progress elsewhere. This method has the disadvantage of large weight and the energy cannot be liberated explosively.

3. Scientists generally agree that if sufficient  $U_{235}$  could be collected together a violent explosion would occur. It has been estimated that 5-10 kgs of  $U_{235}$  assembled with a moderate amount of graphite would yield an explosion of the same violence as 10,000 tons of T.N.T.

4. This Laboratory early became interested in the potentialities of atomic energy and has worked actively in this field since its inception. In the beginning the research centered on production of UF which at that time was a difficult material to synthesize and the lack of which constituted a serious bottleneck in research being carried out by others. By June 1940 it had become clear that the primary barrier to the practical utilization of atomic energy was the enormous problem of separating the Uranium isotopes.

5. At least a dozen schemes have been proposed and investigated as methods for accomplishing the separation. Four of these have been developed to the state where their incorporation into a manufacturing plant is under consideration. These methods are:

- A. Hertz gaseous diffusion
- B. Ultra Centrifuge
- C. Mass Spectrograph
- D. Liquid Thermal Diffusion

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6. A goal of LKG 90%  $U_{235}$  has been established. On this basis the cost of a separating plant employing any of these schemes is over \$25,000,000.

7. This report covers the work which has been done on the liquid thermal diffusion method.

(c) History of Liquid Thermal Diffusion Research

8. All the other methods that have so far proved to be practical for the separation of Uranium isotopes were known previous to September 1939 or have involved a minimum of developmental work in arriving at the present status. Liquid thermal diffusion was considered quite impractical for isotope separation. Indeed, work at four laboratories in this country indicated that gaseous thermal diffusion, a close relative of liquid thermal diffusion, would not even produce a measurable separation. Furthermore, in October 1940, research performed at the General Electric Laboratory in Schenectady on this latter method yielded negative results. It was not until June 22, 1942 that measurements were obtained which proved the method might be of real value.

9. The use of liquid thermal diffusion as a means of separating isotopes was begun by two German scientists, Clusius and Dichei. They discovered, in 1938, that large isotope separation could be obtained by gaseous thermal diffusion. In a short time these men also showed that isotope separation of salts could be made to occur using liquid thermal diffusion and employing water solutions for the working substance. They obtained an exceedingly small separation of the isotopes of hydrogen by using water as a working substance. No work on the subject has been published in this country. The author's interest in liquid thermal diffusion was first aroused in July, 1940 while employed by the Carnegie Institution in Washington. At that time, various means of separating Uranium isotopes were under consideration. A review of the literature showed that among possible methods thermal diffusion had considerable promise, especially because it appeared that considerable quantities of material might be handled by this method. A review of the literature showed that the German scientists had been the principal, if not the sole workers, in the field. Using water solutions of zinc salts they had obtained separation factors of 1.2 (20% separation). However, this separation was accompanied by a seven-fold relative change in the salt concentrations of the two fractions obtained. In other words, one could obtain from the top of the apparatus a solution possessing a separation factor of 1.2 with respect to the bottom fraction. However, the top solution possessed only 1/7 as much solute per cc as the bottom.

10. By expanding this apparatus one could expect to obtain a separation factor of 2, but the top solution would then possess 1/2500 as much salt as the bottom fraction. As one attempted to go to a larger isotope separation, he would find he could obtain such separations but only at the cost of obtaining vanishingly small yields of separated material. The German workers appeared to consider liquid thermal diffusion a scientific curiosity and made no attempt to develop the method into a practical means of separating isotopes. What the Germans have done since September 1939 is a matter for conjecture.

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Since the laws governing the phenomena had not been systematically investigated, a program was undertaken at the Carnegie Institution of Washington to survey the potentialities and laws governing this phenomena. From the beginning, the Naval Research Laboratory was actively interested, and in September 1940 the Laboratory entered into a contract with the Carnegie Institute of Washington providing \$2500 to enable the experiments to be continued and expanded. In October 1940 the experiments were moved to the National Bureau of Standards. At that point Carnegie Institution of Washington was paying the author's salary, the Naval Research Laboratory was furnishing the necessary equipment and the Bureau of Standards was providing laboratory facilities and a chemist who assisted in chemical development. This arrangement continued until June 1, 1941 when the author entered the employ of the Naval Research Laboratory. A report was submitted to the Director of the Naval Research Laboratory describing in detail the results obtained between July 1940 and June 1, 1941. Eleven liquid thermal diffusion columns were constructed. These were made of concentric tubing about 1-1/2 inches in diameter and from 2 to 12 feet long. A number of different wall spacings were used ranging from 2 mm to .5 mm. With these columns water solutions of potassium salts were employed; separation factor and relaxation time were measured as a function of length, wall spacing and temperature difference between the hot and cold wall. The crucial variables proved to be temperature difference and especially wall spacing. This latter variable, when changed by a factor of 2, caused the relaxation time to vary from a few minutes to many hours. It is of interest to note that later research has shown that the findings on water solutions have very limited application to the study of what occurs when  $UF_6$  is used as a working substance. However, the use of salt solutions did point to the variables which might prove important and at the beginning of the research an insufficient amount of  $UF_6$  was available to test the method.

11. After the research on potassium salts was completed, an attempt was made to apply the columns to the isotope separation of water solutions of Uranium salts. This proved to be quite hopeless because all water solutions of Uranium salts tended to decompose at the surface of any practical wall materials. In addition, there was the problem of overcoming the relative change in concentration between top and bottom fractions. As the work progressed it became increasingly clear that to be practical, the working substance in the liquid thermal diffusion apparatus must be a single stable compound. The sole known Uranium compound which might work was  $UF_6$ . However, no supply of this material was available and previously only a few grams of the substance had ever been made. A new suitable method for producing the substance in quantity was devised and a number of kilograms were prepared. Preliminary experiments to test vapor pressure, stability and corrosiveness of the substance were carried out. As a result  $UF_6$  was introduced into two thermal diffusion columns and runs were made. Results which appeared to be positive were obtained but later experiments seemed to indicate that the analysis of the mixtures which was performed by Alfred O.C. Nier and L.F. Curtiss might not be correct. At that time these men were just beginning to make the Uranium isotope analysis and the results were very little different from zero.

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12. About June 1, 1941 the decision was made to pursue the study of liquid thermal diffusion using 36 foot columns at the Naval Research Laboratory. It was felt that a number of columns should be built possessing various spacings and that these columns should be tested at temperatures as high or even higher than the critical temperature of  $UF_6$ . For this purpose a 20 horse power high pressure, gas fired boiler was ordered and ultimately installed. This unit has proved to be capable of delivering 750 lbs. of steam per hour at a pressure of 600 lbs. per square inch. Other materials including a centrifugal pump were likewise ordered. Due to various delays on the part of the suppliers, the equipment was not completed until November 1, 1941. At this point valuable time was lost when an experiment was performed using carbon tetrachloride as a working substance in the column. Positive results were obtained showing separation in the chlorine isotopes and the effect could be easily measured by a change in density which occurred. It was found that the light fraction differed in density by one part in 200 from that of the heavy fraction. However, accompanying the separation was a decomposition of carbon tetrachloride, resulting, probably, in the formation of compounds of the general formula  $C_n Cl_{2n-2}$ . Indeed, in order to analyze the material obtained in the column it was necessary to perform extensive distillations in a fractionating column; ultimately it was found that the column had been ruined since it was impossible to clean out the heavy solid decomposition products of carbon tetrachloride. It was necessary to construct a new column (.53 mm spacing) which was completed about January 1, 1942. This was given necessary preliminary conditioning treatment, to be described later in this report, and placed in operation. The first reports of analysis of samples contained in this device were obtained about February 1, 1942 and gave the first positive results (2% separation). Further experiments with the column yielded a 2-1/2% separation.

13. While these tests were in progress a new column was constructed which employed an increased spacing (.65 mm). It was put into operation March 1, 1942 and numerous runs were made under varying conditions with it. These yielded a maximum of 1.4% separation. Prospects that liquid thermal diffusion might be of value for large scale separation of Uranium isotopes seemed dark indeed at this point. However, a third further column was placed under construction employing a spacing of .38 mm. The first results from this were forth-coming on June 22, 1942. These showed a separation of 9.6% and indicated that by decreasing the spacing between the hot and cold walls to a somewhat smaller value that better results still, might be obtained. One of the encouraging features was the fact that the time necessary to obtain substantially an equilibrium separation was two days. At this point, it is convenient to introduce what we call a pseudo equilibrium time which is defined as the time required for the apparatus to produce a separation equal to 1/2 of the equilibrium value. The pseudo equilibrium time for this new column was 8 hours and in spite of the increased separation that was obtained, this time was not greater than that found in the columns possessing a wider spacing. This finding was in complete disagreement with such predictions as had been made by experts in the field of isotope separation. In fact, one well known expert had predicted a relaxation time for such a liquid thermal diffusion device of 100 years. During July another column was placed in operation having a spacing of .2 mm and this led to a separation factor of 21%. During the construction of the

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pilot plant a further column was tested which possessed a spacing of .14 mm. The results from this apparatus clearly indicated that the optimum spacing was greater than this value and, in fact, separation factor fell to 12.6%. It is felt that while the optimum spacing has been bracketed the best one has not yet been obtained. Indeed, the evidence suggests that optimum spacing is a function of hot wall temperature. Tests are currently in progress to determine optimum spacing and hot and cold wall temperatures using the 36 foot experimental columns and it is felt that the proper operating spacing will be found in the near future.

14. By August 1, 1942 it had become clear that the erection of a pilot plant was desirable. The use of a single column to acquire information required too much time to get all the information that was needed for design of a large plant. Furthermore, it was felt desirable to gain experience in the method of connecting columns in series and to obtain experimental evidence on the pseudo equilibrium time as a function of the number of columns connected in series. In addition, the permissible rate of withdrawal of material was desired. The pilot plant was expected to provide information as to the dependability of the units and the feasibility of operating a large plant with a minimum of difficulty. By Nov. 15 the plant was in a practically completed state and the first tests were begun. Most of the difficulties which required attention arose from such matters as inadequately tightened plumbing, pumps connecting to run backwards and so forth. By December 1 these troubles had been attended to, 5 columns had been cleaned fluorinated and filled with material. In the period between December 3 and December 17 the apparatus was run continuously with no shut down or break down what so ever. Indeed, so constant were the various temperatures and operating characteristics that practically no attention was required to insure successful operation. Many days passed in which operating personnel did not touch any control device such as a valve or variac. No sign has arisen of any defect or difficulty which might cause a progressive deterioration of the apparatus. In fact, it is felt that any breakdown which will occur will be due to established commercial equipment rather than to the columns themselves. The sole difficulty regarding column construction which has arisen to date resulted from an arbitrary decision which was, perhaps, not wisely made. If the o.d. of the monel spacers which center the nickel with respect to the copper is made 3 thousandths of an inch less than the i.d. of the copper at room temperature, then under operating conditions these two will just coincide. However, this arbitrary choice does not allow for the fact that copper is by no means circular. Hence, as the temperature of the copper and the nickel begin to approach operating conditions the monel will tend to bind on the copper. The difference in length between the copper and the nickel changes about one-half inch on going from ambient to operating temperatures. Hence, if a condition arises in which motion is restricted tremendous forces are set up in the tubing. These consist, generally, in tension in the copper and compression in the nickel. In brazing 12 foot sections of copper together annealing of this metal occurs at the junctions. Hence, as the metal is subjected to tensile force the copper tends to give at the point at which it was annealed. Simultaneously, with an increase in length occurs a constriction in diameter which can easily result in the normal spacing of 10 thousandths of an inch being cut to less than 1 thousandth, thus interfering with

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normal operation of the apparatus. The difficulty was easily detected by measurement of the expansion of the nickel relative to the copper on going from ambient to operating temperature and then returning again to ambient temperature. If conditions are proper a relative expansion of approximately 1/2 inch should occur on establishment of operating conditions. On cooling the length of the tubing should return to their initial conditions. If it is found that the copper has assumed a new and longer length the source of the difficulty will be detected. In each of the 3 cases where this constrictive difficulty appeared it was detected before UF<sub>6</sub> was admitted to the column. This difficulty occurred in 3 of 9 columns which have been constructed. In the past on the 36 foot experimental columns 4 thousandths of an inch instead of 3 thousandths of an inch had been allowed between the copper and the nickel and no such difficulty was ever experienced.

15. At present 6 columns are in operating condition and additional units are under construction. Assembly time for a unit is somewhat less than a day and a half. All other features of the plant are completed so that in order to place the 14 units in operation merely requires the completion of 8 additional columns.

THE 36 FOOT EXPERIMENTAL PLANT

(a) Experimental Results

15. In the study of the variables which govern liquid thermal diffusion, one of the most important is the spacing between the hot and cold walls. This variable is the one least susceptible to theoretical study. Experimental results have shown it to be the most crucial of all the variables connected with this effect. The experimental method employed was to construct a column possessing an arbitrarily chosen spacing and then to study the performance of the device keeping the cold wall at approximately 60° and varying hot wall temperature. The following table indicates the nature of the results.

Column No.	Spacing	Amt. of UF <sub>6</sub> in column	Best sep'n. obtained.	Approximate pseudo equilibrium time	Power consumption $\Delta T = 150^{\circ}C$
1	.53 mm	3200 grams	$2.3 \pm .2\%$		16,000 cal/sec.
2	.66 mm	3800 grams	$1.6 \pm .2\%$	8 hours	12,000 cal/sec.
3	.38 mm	2300 grams	$10.6 \pm .3\%$	8 hours	21,000 cal/sec.
4	.21 mm	1400 grams	$20.4 \pm .3\%$	15 hours	32,000 cal/sec.
5	.14 mm	1100 grams	$12.6 \pm .3\%$	13 hours	40,000 cal/sec.

From this table it would appear that the best spacing lies somewhere between that employed in columns 3 and 4, i.e., in the region between the spacing of .21 mm and .38 mm. This best spacing will be, in part, determined by pressure of steam which is available. A description of the results from individual columns follows:

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Column #1

17. This apparatus was the first one constructed and a post mortem investigation showed that the spaces were not as well machined as they could be. In part of the column, the nickel was not constrained in a position concentric with the copper. Furthermore, the isotope analysis performed elsewhere gave trouble. However, a number of runs made at 170°C and 210°C gave a value 2.3% as the best result and indicated a relaxation time of about one day.

Column #2

18. In studying this apparatus improvements were made in sampling technique and the method of sample withdrawal standardized. In all cases, 90 grams of material were removed from the column in such a way as to create a minimum disturbance to the distribution of material.

Hot Wall	Cold Wall	Length of Run		% Separation
170°C	62°C	20	hrs.	1.2 ± .2%
215°C	62°C	24	hrs.	1.1 ± .2%
190°C	62°C	27	hrs.	1.3 ± .2%
150°C	62°C	27	hrs.	1.2 ± .2%
190°C	62°C	8-1/2	hrs.	.7 ± .2%
190°C	62°C	2	days	1.3 ± .2%

19. A rate of flow experiment was made at 190°C hot wall, 62°C cold wall. This experiment showed that if one had two of this type connected in series and introduced ordinary UF<sub>6</sub> at the neutral point between them, UF<sub>6</sub> could be withdrawn from the top of one and from the bottom of the other possessing a separation of 1.4% and at the rate of 2 kilograms in each fraction per day. A similar experiment showed that at a hot temperature of 150°C, employing two columns in series one could get a separation factor of  $3.2 \pm .4\%$  and withdraw material at the rate of 2 kg of each fraction a day. This result indicates that with a wide spacing lower hot wall temperatures are probably advantageous. This is perhaps due to parasitic convection currents which can occur when wide spacings are employed. The figures quoted point to pseudo equilibrium on time of about 8 hours for column 2.

Column #3

20. The experiments performed on this apparatus showed it to be far superior to the previous columns. The following results have been obtained.

Hot Wall	Cold Wall	Length of Run		% Separation
170°C	64°C	1	day	8.6%
170°C	64°C	2	days	9.2%
170°C	64°C	4	days	9.6%
210°C	64°C	22	hrs.	8.6%
210°C	64°C	2-1/2	days	11.6%
210°C	64°C	4	days	10.6%

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In these experiments the samples taken off were 100 grams each. If there is no error in the experiment or analysis, a pseudo equilibrium time of less than a day - of the order of 10 hours at a hot wall temperature of 170° is indicated. Pseudo equilibrium at the higher temperature appears to be longer - about a day, on the basis that figure 11.6% is correct. However, there is an inconsistency in the figures for 2-1/2 days and 4 days, which casts doubt on the reliability of 11.6% result. If the 10.6% value is correct then the relaxation time at 210°C is about the same as at 170°C. Flow experiments have been made showing that one can withdraw material in two fractions possessing a separation of 7% and at the rate of 1 kg of each per day.

21. The improvement of Column #3 over Column #2 becomes clear when it is recalled that virtue of a separation apparatus is measured by  $(\log S)^2 F$  where S is the separation factor and F is the rate at which each fraction can be removed. On this basis, Column #3 is as much as ten times as good as Column #2.

Column #4

22. The experiments on this apparatus did not yield the marked improvement over Column #3 that was anticipated. Nevertheless, an improvement was obtained:

Hot Wall	Cold Wall	Length of Run	% Separation
170°C	64°C	1 day	7.8%
170°C	64°C	2.2 days	13.0%
170°C	64°C	4.2	19.0%
210°C	64°C	1 day	13.5%
210°C	64°C	2.5 days	18.2%
210°C	64°C	4.5	20.4%

23. In the case of this apparatus 55 gram samples were withdrawn. Consideration of the hold-up of the column and its pseudo equilibrium time shows that two fractions of  $U_6^{235}$  700 grams each can be withdrawn from this column, per day possessing a separation factor of 14%.

24. The figures indicate an improvement over Column #3 in  $(\log S)^2 F$  of a factor of 2.8.

Column #5

Hot Wall	Cold Wall	Length of Run	% Separation
170°C	60°C	1 day	1.7%
170°C	64°C	3 days	5.6%
215°C	57°C	1 day	8.4%
215°C	57°C	2 days	11.1%
215°C	57°C	3 days	11.7%
230°C	60°C	1 day	9.8%
230°C	60°C	2 days	12.6%

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25. At this point it should be emphasized that when we speak of hot wall temperature of 170° we mean that saturated steam at a gauge pressure of 100 lbs is in contact with the inside of the nickel. In actual fact a film of water will at all times be found upon the surface of the nickel and a certain temperature difference will exist between the outside of this film and the nickel wall itself. In addition, there is a temperature drop through the nickel. In the case of very small spacings the temperature drop in this water film and in the nickel may come to be a considerable fraction of that in the UF<sub>6</sub> itself. Our measurements show that UF<sub>6</sub> has a thermal conductivity of the order of  $4 \times 10^{-4}$  calories per degree centimeter. A similar comment holds true in the case of the cold wall. In the case of the cold wall, water at a temperature of, say 64°, was caused to circulate past copper at the rate of about 10 feet per second. At the surface of the copper is a thin film of water which is motionless. For the heat to be conducted through this film a gradient of temperature must exist and as a consequence it would not be surprising if the temperature at the inside circumference of the copper were 10° higher than that of the circulating water. It would appear that the foregoing phenomena constitutes a large part of the reason for the decrease in separation which was observed in Column #5. It is readily seen that in the limit as the wall spacings approaches 0 that all the temperature drop will occur in the two surface films of water and in the copper and nickel.

(b) Operational Characteristics

26. Throughout the work on the 36 foot columns careful attention has been paid to determining the dependability of the columns. If a large plant is to be set up capable of producing 1 kg of 90% U<sub>235</sub>, many thousands of units must be employed. This will be true of any device which has, to date, been employed for isotope separation. Unless each individual unit which is to make up the total pyramid is absolutely dependable and free from all troubles, the difficulty of operating the assemblage quickly becomes astronomical. To be practical the device must be capable of a trouble-free operation which is at least equivalent to the performance of electrical motors. Indeed, it would be desirable to surpass even their splendid degree of perfection. Naturally, in the time which has elapsed it has not been feasible to subject our columns to long time performance tests, the maximum length of time for which any column has been in operation is 500 hrs. However, on the completion of experiments, the experimental units have been completely dismantled and parts carefully examined for signs of corrosion or of any progressive wear whose cumulative effect might tend to impair operation of the column. Furthermore, the possibility has been considered that UF<sub>6</sub> might undergo dissociation at the temperatures at which we work. No evidence for this effect has been found. After operating columns for 500 hours the nickel which forms the hot wall was removed and inspected. It was found to be entirely unchanged, presenting the same shiny appearance that it possessed when the column was initially assembled. These observations give rise to the belief that thermal diffusion columns can readily be made to possess an unusually high degree of operating dependability.

THE PILOT PLANT

(a) Design

27. Design of the plant was considerably influenced by the need for

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speed. For instance, because of the availability of a heating boiler, the decision was made to use this source of power in spite of the fact that the maximum pressure obtainable from this device was 165 pounds per square inch. Our experience in the past has indicated that a considerably higher pressure is desirable. The length of the columns was set at 48 feet, largely because existing facilities would conveniently permit of this arrangement. It was felt that greater length was desirable (192 feet). However, in the interest of speed, the decision was made for the shorter length.

28. Many of the other design characteristics of the columns are quite arbitrary: the diameter of the inner nickel tubing which acts as the hot wall was chosen to be 1-1/4 inch i.p.s. (1.667 inch o.d.); the factors which set this diameter were these: a smaller diameter would result in lessened production per column, accompanied with a severe increase in the steam pressure drop within the column; a larger diameter, say twice as great, would increase production by a factor of 2 at most. However, if the wall thickness were maintained constant the rigidity of the tubing would decrease to 1/8 and even if the wall thickness were increased to compensate for the increased diameter the variation in diameter of the tubing would still be much larger than in the case of the 1-1/4 inch i.p.s. It would appear that some increase (25-50%) in diameter might be feasible. However, since our earlier experience was with tubing of 1-1/4 inch i.p.s. it did not seem advisable to introduce a new experimental variable in view of the slight advantage to be gained. The choice of nickel as the hot wall metal was made because of the experience that nickel is probably the least corroded by  $UF_6$  of any elements in the periodic table. A possible substitute for nickel is aluminum or some of the aluminum alloys such as duralumin. Corrosion tests have shown these materials to be very resistant to  $UF_6$  and were the shortage of nickel to be severe it is probable that these materials could be used.

29. The material chosen for the cold wall was copper. At the operating temperature copper is practically unaffected by  $UF_6$  provided a preliminary fluorination has provided a protective fluoride coating on the metal. Other considerations included is the fact that copper is a good thermal conductor and that commercial copper tubing presents a surprisingly good uniformity of diameter and roundness. It is likely that any one of many metals could be used for the cold wall. The wall spacing between the nickel and copper tubing was chosen on the basis of experimental results on the 36 foot test columns. Under operating conditions this distance is .25 mm. Accordingly, the i.d. of the copper was set at 1.690 inches and an o.d. of 2 inches was arbitrarily selected.

30. With the spacing (.010 inches) which in operation exists between the inner nickel and outer copper walls, the question naturally arises as to the importance of exactly maintaining this dimension. Commercial tubing possesses deformities or irregularities which can cause a deviation from the chosen dimension, for instance copper tubing varies in its i.d. due to ovality from 1.688 to 1.692. This would mean a spread of from 8 thousandths to 12 thousandths in the spacing. While this variation extends over only a short section of the tubing a spread of from 1.689 to 1.691 is quite common and it is rather fortuitous to find tubing which does not deviate to some extent from the average i.d. of 1.690 inches. In addition, the copper tubing is never

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straight. There are always bends and kinks. Likewise the nickel tubing possesses ovality and is not always straight. In selecting the tubing that was used, ordinary commercial copper was obtained. In view of the large tolerances which the International Nickel Company prescribes for its tubing, we asked the company to supply us material which would conform to somewhat better specifications. This the company was readily able to do. With lengths of tubing which are employed it seems necessary to employ spacers in order to center the nickel with respect to the copper. It seems obvious from the consideration of the straightness of the tubes that these spacers must occur at least every 2 feet. To date, the method used for fastening the spacers has involved the drilling and tapping holes in the nickel and screwing in 5-40 monel studs into the nickel. Subsequently, the tubing is placed in a lathe and the monel is turned down to the proper diameter. Various frequencies of spacing are employed, but in all cases a set of spacers consists of 4 placed around the nickel tubing at 90° intervals. The distance between sets which has been employed have been 6 inches, 12 inches and 24 inches. We have not yet determined the optimum distance between spacers, but experiments are in progress. It seems probable that, as previously mentioned, there must be at least one set every two feet. From the standpoint of labor alone, it seems desirable to maintain the frequency at less than one set every 6 inches and, in fact, a greater frequency might even be harmful to the process since the spacers must produce a local non-uniformity of the heat gradients. Using the 36 foot column, an experiment was inadvertently conducted in which spacers did not center the nickel within the copper and isotope separation did occur. We are under the impression that the successful working of a column is not largely interfered with by a lack of centering of the tubing or by ovality of either the copper or the nickel.

31. In the pilot plant a closed steam system is employed. Steam is conducted from a heating boiler to a manifold which is in the vicinity of the top of the columns. This manifold possesses 14 risers each serving a single column. At the bottom of the columns a similar manifold is located. Each column possesses a steam valve at its top and bottom. Steam is fed in at the top for obvious reasons and condensate collected in the manifold at the bottom. This condensate is lead through a trap to a condensate return pump which feeds the water back into the boiler. To date, the steam operating pressure employed has been 100 pounds.

32. Each individual column has its own water circulating system in which water at approximately 50° is kept in motion by a Worthington 4 DE 15 horsepower centrifugal pump. Under our conditions this pump causes a flow of approximately 1,000 g.p.m. On this basis the temperature difference between the bottom and the top of the column is .5 of a degree C. To maintain the temperature at 50°C hot water is bled from the top of the system and 9 g.p.m. of cold water (5°C) is fed in at the suction side of the pump. Choice of centrifugal pump was arbitrary and there is no reason to believe that a circulation of 50 to 100 g.p.m. would not be adequate. Experiments will be made to determine the effect of such a pumping speed.

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(b) Materials

<u>Name</u>	<u>Weight</u>	<u>Cost</u>
672 ft. of 1-1/4 inch i.p.s. nickel tubing	1,750 lbs.	\$ 1,672.00
672 ft. of 2 inch o.d. copper tubing	2,000 lbs.	540.00
1500 ft. of 4 inch i.p.s. iron pipe	18,000 lbs.	1,800.00
900 ft. of 2 inch i.p.s. iron pipe	4,000 lbs.	500.00
assorted 5 inch and 6 inch iron pipe	4,000 lbs.	400.00
structural steel	5,000 lbs.	600.00
brass and bronze valves and fittings	400 lbs.	
miscellaneous copper	200 lbs.	
miscellaneous nickel	100 lbs.	
15-4 DE centrifugal pumps at 700 lbs. each		
10,500 lbs. mainly iron at a total cost of		5,400.00

(c) Cost of Operation

33. The plant, to date, has been operated with steam pressure at 100 lbs. per square inch and cold water temperature at 50°C. Under these conditions 100 B.T.U. per second of steam heat is consumed and 9 g.p.m of cold water at 5° is required per column. This steam consumption is approximately 450 lbs. per hour per column and with our boilers we find that 1250 lbs. of coal are used each day. At \$5 a ton for coal this amounts to \$3.15 per day. Cost for cold water is \$.25 per column. At \$.01 per kw hr the power charge per column for the circulating system is \$2.50, giving rise to a total cost exclusive of operating manpower of about \$6 a day per column. On a large scale with favorable location relative to a coal mine the steam cost would decrease to about \$2.50 per day and it is quite probable that charge arising from a circulating system might be reduced to less than \$1 per day, giving rise to a total of \$3.50 per day per column.

(d) Operating Methods and Techniques

34. We shall consider first the testing and operation of a single column. The techniques that are used in operating the column hinge to a large degree on the physical and chemical properties of UF<sub>6</sub>. As has been mentioned previously, in this report, considerable research has been devoted to determination of the physical and chemical properties of this substance. Thus, it is the chemical properties of UF<sub>6</sub> which determines that nickel must be used with the hot wall and that no organic material (in the form of packing) may be used in this device. Furthermore, at the temperature at which it is desirable to operate, the compound possesses a vapor pressure of about 400 lbs. per square inch. Accordingly, in construction welded and brazed joints must be tested hydrostatically to insure that there should be no leak whatsoever. The component parts are tested to a pressure of 800 lbs. per square inch. Upon completion of the column and after it is fastened into its operating position, procedures are carried out which remove any excess oil from the working space. These procedures consist of washing with carbon tetrachloride and the passage of high pressure hot water through the column. The device is then thoroughly

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dried and fluorine is introduced into one end of the column which is heated to 120°C. This stream of gas is maintained until fluorine is observed to appear at the opposite end of the apparatus which generally requires about 2 hours when the flow of gas is 2 CC per second. The action of the gas is to form a protective coating on the nickel and copper and to remove any last traces of organic materials or other substances which might cause decomposition of the  $UF_6$ . The apparatus is then ready to be filled with  $UF_6$ . Water is caused to flow in the circulating system and high pressure steam is admitted to the inside of the nickel. The storage chamber containing somewhat more than enough material to fill the column is connected to its bottom and the temperature in the chamber is increased until it is approximately 20° above that of the hot wall of the column. Under these conditions the vapor pressure of  $UF_6$  in the storage chamber drives the material into the column and maintains it there. In running, the storage chamber is left connected to the column and is maintained at the 20° higher temperature. Thus provision is made against the creation of excessive hydrostatic pressure within the column. Were the storage chamber to be disconnected and the  $UF_6$  maintained in the column without a provision for an exit, then an increase in either the steam pressure or water circulating temperature would be accompanied by an increase in volume of the  $UF_6$ . An excessive cooling would also be harmful since then proper pressure would not be maintained on the column. Ultimately, if the temperature were too low the material would solidify on the cold wall. Finally, when too much of the material is solidified very peculiar things happen in the column and the remaining liquid material therein pitches violently. Keeping the storage chamber connected to the column thus maintains a constant pressure upon the material and provides against drops which otherwise occur when inevitable temperature variation comes.

35. To date it has not been found necessary to use any valves or other moving parts in the column. A valve has been developed and tested which is proving satisfactory, but as yet has not been used largely because it was unnecessary to employ in the present plant. In removing samples 1/4 inch monel tubing (1/8 inch i.d.) is employed, this tubing taps into the column at the top and at the base and extends out approximately 5 inches. Under normal operating conditions this tubing is filled with  $UF_6$  which, at the end away from the column, is solid. To draw off material from the column, a container is fastened to the monel tubing. Monel is then gently flamed, the chunk of solid material melted and the container is practically instantly filled with  $UF_6$ .

36. In shutting down or stopping operations the method usually employed is to freeze the tubing connecting storage chamber and the column. Steam is then cut off and when the column is cool circulating water is stopped. This method prevents an excessive amount of  $UF_6$  from flowing in while the column is cooling. The column possesses the proper amount of material so that when running condition is reassumed very little flow of material out of or into the storage chamber is necessary.

37. A simple means has been devised and used to make possible connecting up a number of columns in series. A convective loop is established between the top of one column and the bottom of the next. In this

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loop, which consists of 1/16 inch i.d. tubing, material goes up one tubing and down the other. Motion is established by subjecting one tubing to a temperature of approximately 150° and the second to a temperature of approximately 110°. Density of the material in the tubing at 150° is .2 -.3 grams per cm<sup>3</sup> less than that in the cooler tubing. As a result, material flows down in the cool side of the loop and up in the hot side. A total driving force around the loop of about 5 lbs per square inch is obtained. It is obvious that this driving force could readily be increased by changing the two temperatures, and, in fact, were it necessary a driving force of as much as 30 lbs. per square inch could be obtained. However, viscosity of UF<sub>6</sub> at 130°C is of the order of one centipoise and a satisfactory circulation is obtained under the conditions which have been discussed. This method of circulation has been used and proved to be quite satisfactory. To date, 4 columns have been connected in series with satisfactory results.

(e) Experimental Results

38. Sufficient time has not elapsed since the beginning of operations on the pilot plant to accumulate many experimental results. It has been thought best to start work by testing individual columns first. Then individuals have been connected to form series pairs.

39. One such pair gave the following results:

Time	Bottom Sample	Top Sample
1 day	-10 %	3.8%
3 days	-20.9%	11.2%
5 days	-26.5%	15.0%

40. These were obtained using a hot wall temperature of 170°C and a cold wall temperature of 50°C. Previous experience has shown a more rapid approach to equilibrium when higher operating temperatures are employed and this is expected to be also true in the case of these columns. One important consequence of this experiment is the light it sheds on the effect of dead space at the top of the columns. There is little doubt that the slower rate of approach to equilibrium at the top of the second column of the pair is due to this dead space. It seems feasible to diminish this volume by more than 100 cc. by a re-design of the top siphon assembly. This change can be accomplished in units which are not about ready for assembly and will not result in a delay in the testing of the plant.

COSTS OF LIQUID THERMAL DIFFUSION RESEARCH

41. To date some \$64,000 has been expended on liquid thermal diffusion research. The amount of money spent on materials between July 1940 and June 1, 1940 was \$1100. In that period salaries of \$5000 were chargeable to the work. Between June 1, 1941 and July 1, 1942, a period which covers most of the work on 36 foot units, the sum of \$18,000 was expended. This included all salaries, materials and overhead chargeable to the project. Between July 1, 1942 and December 1, 1942 \$40,000 was expended - principally for the pilot plant. A breakdown of this figure follows:

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Materials: tubing, pipes, valves etc.	\$ 7,000
Centrifugal pumps	5,400
Conversion of Heating Boiler	9,000
UF <sub>6</sub>	3,000
Salaries of scientific personnel	5,000
Salaries and overhead of shop labor	10,600
	<hr/>
	\$40,000

RESULTS FROM THEORETICAL STUDIES

42. A theoretical study of liquid thermal diffusion has been begun by Dr. Rosen of the University of North Carolina. Calculations have already been made to determine the number of units required for 1 kg of 90% U<sub>235</sub> per day. The unit taken is assumed to consist of two 48 foot columns connected in series. Material would be fed in at a point between the columns. Enriched material would be withdrawn from the top of one column and impoverished substance from the bottom of the other. The enriched material would form part of the feed for the next layer and the impoverished substance would form part of the feed for the preceding layer. The output per unit is taken as 1 kg. UF<sub>6</sub> per day and the number of columns required is calculated a function of S. In this connection S is defined as the isotope ratio between the input and the enriched materials. The results of two of these calculations are shown in the following tables. The units required are given in round numbers. In the case of S = 1.50 the stripping section is included. In the case of S = 1.1 additional stripping is needed requiring about 5000 additional units.

Objective:	1 kg 90% U <sub>235</sub> per day
Separation:	S = 1.50
Transport per Unit	= 1 kg per day in positive direction

Stage	Units in parallel	% U <sub>235</sub> of Feed
n = 0	419	.71
1	281	1.06
2	189	1.53
3	127	2.36
4	86	3.49
5	59	5.15
6	41	7.53
7	28	10.9
8	20	15.5
9	16	21.6
10	11	29.2
11	9	38.2
12	7	48.1
13	6	58.2
14	5	67.6
15	4	75.8
16	3	82.4
17	2	87.6
Total Number	1312	Output 91.4 % U <sub>235</sub>

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Objective: 1 kg 90% U235  
Separation: S = 1.10  
Transport per Unit = 1 kg in Positive Direction

Stage	Units in parallel	% U235 of Feed
n = 0	2092	.71%
1	1902	.78%
2	1731	.86%
3	1575	.94%
4	1433	1.04%
5	1305	1.14%
10	915	1.82%
15	512	2.90%
20	323	4.59%
25	206	7.19%
30	134	11.1 %
35	88	16.7 %
40	50	24.4 %
45	42	34.3 %
50	31	45.6 %
55	23	57.5 %
60	16	68.5 %
65	13	77.8 %
70	7	85.0 %
74	2	89.2 %
Total Number	23,800	Output 90.1 %

43. Other characteristics of the units are being investigated including the relaxation time of both the pyramid, and individual columns under varying conditions. An attempt will be made to develop a theory of the processes by which isotope separation is obtained.

FUTURE UNITS

44. Previously in this report the reasons for selecting a 48 foot length for the column have been detailed. Consideration of the factors that go into successful operation of the column show that a more practical length for large scale operation would be approximately 192 feet. Our unit would consist of two 192 foot columns connected in series. A reasonable estimate of the performance of such a unit can be made. This is based on the assumption which is backed by experimental evidence that separation factor increases as  $e^1$ . On this basis an equilibrium separation factor  $S = 7.4$  would be obtained between the ends. This corresponds to introduction of material at .71% U235; equilibrium samples containing 1.92% U235 could be taken from the top and samples containing .26% U235 taken from the bottom. It should be emphasized that this represents the equilibrium value and that in the operating state one would run at operating conditions in which material was removed at 1.1% and injected at .46%. It will be possible to obtain a fairly good idea of the pseudo equilibrium time and output of such a unit by a test employing present units of the pilot plant.

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POTENTIALITIES FOR IMPROVEMENT OF APPARATUS

45. The potentialities for improvement of the performance of apparatus have not been exhausted. Optimum spacing has probably not been achieved. In view of the crucial nature of this variable a substantial improvement may be attainable. An improvement in performance will be obtained by the use of better centering of the hot wall with respect to the cold. To date, no real attempt has been made to insure that the two walls should be accurately concentric. Likewise the proper frequency of spacers is unknown.

46. Optimum hot and cold wall temperatures have not been established. No systematic investigation has been made of the effect of varying cold wall temperatures. All indications point to the value of increasing the hot wall temperature. To date, the maximum temperature at the hot edge of the  $UF_6$  has not exceeded  $200^{\circ}C$ . No reason can be seen why this temperature could not be increased to  $300^{\circ}C$  or  $400^{\circ}C$  with advantageous results. The diffusion process would then operate on a combination of liquid and gas with the density of the gas being in the neighborhood of  $1.6 \text{ grams/cm}^3$ .

47. Redesign of the top of the column can cause a decline in pseudo equilibrium time. The use of higher temperatures will also aid in this direction. Attainment of higher separations would probably be accompanied by a decrease in the pseudo equilibrium time for the pyramid.

COMPARISON OF VARIOUS SEPARATION METHODS

48. The secrecy which has been maintained around progress of other methods makes a quantitative comparison between the schemes somewhat difficult. However, it is quite feasible to make a qualitative description of the relative merits of the various methods. The gaseous Hertz diffusion method possesses the following characteristics. The separation factor obtainable in a single unit is low, in the neighborhood of 1.004. Thus, in order to obtain a 90%  $U_{235}$  by this device, it is necessary that approximately 2000 units should be connected in series. The virtue of the apparatus would seem to lie in the fact that large quantities of material could be processed by increasing the area of the diffusing screens. In addition, the time necessary to establish an initial equilibrium could be made reasonably short, perhaps 90 days. By varying the size of the unit according to its place in the pyramid it would be necessary to have only approximately 2000 units. It should be noted, however, that all the units must be connected in series and that elaborate precautions must be taken to bypass units which might fail. Furthermore, one must have these 2000 units whether he wishes to withdraw 1 kg or .1 kg  $U_{235}$  per day. The flow-through centrifugal method possesses a higher separation factor ( $s = 1.02$ ), this figure can conceivably be increased. Estimates have been made for the total number of units having a 1 kg of 90%  $U_{235}$  per day and these involve the building of approximately 5000 of the units. This method produces very large quantities of separated material per unit and again has a virtue of reasonably short relaxation time. However, the operation of an ultra centrifuge at 500 r.p.s. presents obvious technical problems for long time operation. Both the Hertz and centrifugal methods involve exposing moving parts to  $UF_6$  and the use of packing materials

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to prevent the inward motion of air or outward motion of  $UF_6$ . In view of the great chemical activity of  $UF_6$  it would seem debatable whether these devices could give dependable operation for month after month. The third method is the mass spectrograph. The virtue of this device lies in the large separations which can be obtained in one operation. Theoretically, 90%  $U_{235}$  might be produced in one operation. Actually, however, this would involve a prohibitively precise and narrow adjustment of a slit system and would result in negligible yields of material. In practice, a ten fold increase in the purity of  $U_{235}$  can be made at one step. The mass spectrograph has the further virtue that relaxation time is extremely small since the process occurs almost instantly. Units of the mass spectrograph may be operated independently and hence failure of one unit does not react upon the operation of the others. The weakness of mass spectrograph lies in the small yields of material which can be obtained per day. It is probable that these are in the neighborhood of 10 to 100 mg of ten fold enriched uranium per day. A further disadvantage lies in the fact that only a small fraction of the initially volatilized material will reach the collecting plates. This is not serious as long as the initial material is .7%  $U_{235}$ . However, if one were to attempt to reprocess 7%  $U_{235}$  the difficulty becomes tremendous. Thus, in order to obtain 70%  $U_{235}$  in two steps with the spectrograph one would face the following dilemma: of the 7% material introduced into the spectrograph probably of the order of 1% would reach collecting surfaces. The remaining extremely costly material would be scattered all over the apparatus. It would be necessary to tear down the equipment and clean all surfaces carefully to recover this precious Uranium which would then have to be reconverted chemically into a form suitable for ionizing. This process would have to be repeated many times before all the 7% material could be processed.

49. Despite the fact that Hertz diffusion and the centrifugal method possess the capacity for large quantity separation, these methods must be made extremely dependable in order to compete with the spectrograph. Indeed, the dependability must be at least of the order of that of a commercial electrical motor, if not greater. While it is difficult to estimate the cost of the number of units necessary to produce 1 kg of 90%  $U_{235}$  per day using the spectrograph it seems likely that the cost of such an arrangement would be well in excess of \$50,000,000. This is also true of the Hertz diffusion and the centrifugal methods. At this date, our estimates place the number of 384 foot units (two 192 columns in series) at \$5250 in order to obtain 1 kg 90%  $U_{235}$ . In order to reach the 90% value 17 must be connected in series. The output of the individual units would be 250 grams of  $UF_6$  per day. A pyramid started with one unit in the top would spread to 250 units at the bottom. Thus, it would be possible in attaining 1 kg 90%  $U_{235}$  to operate 6 separate pyramids possessing 875 units each. These pyramids would be completely independent of each other and individual failure in one would not affect the operation of the remaining. It is estimated that these units could be built on a large scale for \$5000 each, making a total of \$26,000,000. At present, the relaxation time for a pyramid looks to be about 300 days, but our experiments have shown that this time can probably be reduced.

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METHODS OF ARRIVING AT A PRODUCTION RATE OF 1 KG 90% U235 PER DAY

50. To date, all the methods of isotope separation are actually in the laboratory state of development. Were war time conditions not prevailing it is extremely doubtful if any responsible scientist or engineer would recommend the jump to manufacturing scale production. Very slight errors in the design of a unit and slight imperfections in its operation could lead to difficulties of an extreme nature when an attempt was made to operate thousands simultaneously. Indeed, it is not difficult to visualize a state of affairs in which operation of an apparatus might require more trained scientific and engineering personnel than is to be found in these United States. The question arises as to whether the goal of 1 kg 90% U235 is not an excessively high one and whether an approach to that goal should not be made more gradually. It seems hardly possible that any method could be made to begin operation on the 1 kg per day basis sooner than 4 months and this would only be possible through the use of a super-priority on materials and, indeed, most of the methods would require from 8 months to a year before they would be ready to start operations. Upon the passage of the necessary relaxation time material could be withdrawn at 1 kg per day provided none of the usual "bugs" had been uncovered. In approximately a week enough material would be accumulated to obtain the desired reaction. Suppose, on the other hand, that the goal were set at .1 of a kg 90% U235 per day. In the case of the centrifuge, mass spectrograph, and liquid thermal diffusion, the number of units that are required would be approximately .1 that of the larger scale plant and the construction would involve the use of approximately .1 as much strategic materials and manpower. Since a fewer number of units would be required, construction time would be lessened and the difficulties of obtaining materials decreased. In setting up a plant the troubles found in obtaining dependable operation would be cut, resulting in the saving of months of time. If a 1 kg per day plant could be put in operation in as little as a year and 3 months then a .1 kg plant could certainly be placed in production in a year. After the passage of 70 days enough material would be accumulated to produce the desired reaction. It would seem that under even the most favorable conditions a 1 kg per day plant could not be made to produce the initial desired amount faster than a .1 kg per day plant.

AN ALTERNATIVE TO THE 1 KG PER DAY PLANT

51. Such is the extrapolation that is being made in jumping from laboratory conditions to the manufacturing conditions that the question even arises whether haste might not be made more rapidly by decreasing the immediate objective still further.

52. Such a decrease in objective would be practical if a collaboration could be arranged with the group working at Chicago. Theoretical physicists connected with this group have estimated that if 250 - 1000 kg of 1.4% U235 were assembled with suitable graphite a divergent chain would result. This would provide a source of energy whose dimensions would be fairly consistent with naval needs. It is not thought that a really violent explosion would result from the chain. A product of the divergent

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chain is neptunium (element 94) of mass 239. Measurements have demonstrated that a sufficient accumulation of this material (5-10 kg) could be used as a substitute for U235. Neptunium has properties which permit of its easy chemical separation from other elements including Uranium.

53. The goal of 500 kg of 1.4% U235 is one which is capable of relatively easy attainment. On the basis of the use of 192 foot "units" having an output of 250 grams UF<sub>6</sub> per day with a separation of S = 1.50 between input and output, an estimate can be made of the number required. A number of independent pyramids would be used having the following characteristics:

Stage	Units in Parallel	% U235 of Feed
N = 0	4	.71%
1	1	1.07%
Input 1.18 kg	.71% U235	
Output .25 kg	1.60% U235	
Reject .93 kg	.47% U235	

54. A plant consisting of sixty independent pyramids (300 units) would produce 500 kg 1.60% U235 in 82 days of running. Of this time 32 days would be required to allow the plant to attain its initial half equilibrium time state.

55. An alternative arrangement would consist in operating all 300 units independently. The output would be 50 kg of 1.07% U235 and 75 kg 1 day of .47% U235. Since only 16 days would be required for the attainment of half equilibrium, in 82 days after the start of operations a total of 3,300 kg of 1.07% U235 would be produced.

56. An important factor to consider is the purity of the product obtained from our separation method. The diffusion apparatus introduces no impurities. After separation has been performed it is feasible to attain a high degree of purity by use of a fractionating column. Material treated in this way could be converted to very pure Uranium and much of the costly purification currently required for Chicago materials would be eliminated.

57. The estimate of the amount of material needed by the Chicago group to produce a chain reaction is necessarily very crude. Information concerning the many experiments performed by these workers in the last six months has been denied to us. It is vitally necessary that there be an exchange of technical information if proper plans are to be made for future plants and progress of the work is not to be unduly impeded.

CONCLUSIONS

58. Liquid thermal diffusion has been shown to be a practical means of separating Uranium isotopes. The virtues of the method lie in ease of maintaining a pure product combined with a fairly large separation

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per column and moderate output. The principal weakness is high consumption of energy in the form of steam. Experiments employing a unit consisting of two 98 foot columns yielded samples possessing an isotope separation of 45.5% ( $S = 1.455$ ). The unit is capable of producing 1 kg each of two fractions differing in isotope separation by 20%.

59. Potentialities for improvement of the apparatus have not been exhausted. The changing of experimental variables may yet lead to a substantial improvement in performance. Variables to be investigated include: spacing and centering of hot wall with respect to cold wall; cold wall temperature and hot wall temperature.

60. On the basis of 384 foot units (consisting of two 192 foot columns in series), an estimate has been made of the number of units required for 1 kg of 90% U235 per day. Such a plant would require 5250 units costing about \$26,000,000.

61. This 1 kg per day plant compares favorably in cost and probable operating dependability with other proposed schemes.

62. The question is raised whether a 1 kg per day plant is a desirable immediate objective. The alternative of producing either 1.6% U235 or 1.07% U235 to be used in an experiment similar to that in progress at Chicago has been considered and found attractive. For this purpose a plant of 300 units would be sufficient.

## RECOMMENDATIONS

63. It is recommended that the current obstruction to transfer of information between this Laboratory and Chicago should be removed.

64. It is recommended that investigations be continued at this Laboratory employing both the pilot plant and the 36 foot experimental unit. It is felt that three months of further investigations is needed before an attempt should be made to design a larger plant.

## APPENDIX

### (a) Some Physical and Chemical Properties of UF<sub>6</sub>.

65. Once the decision had been made that UF<sub>6</sub> was probably the only Uranium compound which can be used in the liquid thermal diffusion it became clear that a greater knowledge of this compound was needed. In the first place, at the beginning of this research perhaps no more than 100 grams of the substance had ever been produced anywhere before. There had previously existed no reason for a detailed investigation of the compound. The first problem that had to be met was the production of the compound in large amounts. A solution was discovered which today is used commercially for the production of UF<sub>6</sub>. Briefly, this consists in the preparation of UF<sub>4</sub> by an ordinary wet chemical method. This compound is then completely dried and converted to UF<sub>6</sub> by fluorination at 350°C. A

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second problem was the question of suitable containers for the material. It was quickly learned that  $UF_6$  is extremely reactive chemically. It is decomposed almost instantly by water. It reacts with almost all organic materials. In this respect it is closely akin to fluorine. Fortunately, because of a protective coating mechanism the substance can be exposed to certain materials for long periods of time without appreciable decomposition. An investigation was made using a dozen different metals to determine which was superior. It was determined that best performance was obtained if the material to be tested was first heated and exposed for a period of time to the action of fluorine gas. Under these circumstances it was found that at a temperature as high as  $350^{\circ}C$  nickel undergoes practically no corrosion by  $UF_6$ . It was further found that traces of moisture seemed to decompose the protective fluoride coating in the case of some substance. If  $UF_6$  is completely freed from moisture silver, solder is very little corroded at  $150^{\circ}C$ . However, in the presence of traces of moisture, the solder acts like a different substance and is readily corroded.

66. Search of the literature revealed that a crude melting point determination had been made on  $UF_6$  and that vapor pressure measurements had been carried on only to pressures of two atmospheres. It seemed necessary to redetermine the melting point ( $640^{\circ}C$ ) and to carry the vapor pressure measurements up to much higher pressures.

67. The results of these measurements are given in the following table:

Vapor Pressure of  $UF_6$

<u>Temperature</u>	<u>Pressure</u>
$56^{\circ}C$	15 lbs/in <sup>2</sup>
$65^{\circ}C$	23 lbs/in <sup>2</sup>
$90^{\circ}C$	37 lbs/in <sup>2</sup>
$100^{\circ}C$	63 lbs/in <sup>2</sup>
$118^{\circ}C$	97 lbs/in <sup>2</sup>
$137^{\circ}C$	133 lbs/in <sup>2</sup>
$162^{\circ}C$	220 lbs/in <sup>2</sup>
$200^{\circ}C$	440 lbs/in <sup>2</sup>
$230^{\circ}C$	720 lbs/in <sup>2</sup>

The measurements have a probable error of 5%.

The critical point is another physical constant whose value is of interest in connection with this problem. A determination has been made and results in a value  $232^{\circ}C$ . Another set of data which is of importance in arriving at some understanding of the processes of liquid thermal diffusion and in dealing with  $UF_6$  in the liquid state are measurements of density of the substances as a function of temperature. A table follows giving this relation:

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Density of  $\text{UF}_6$

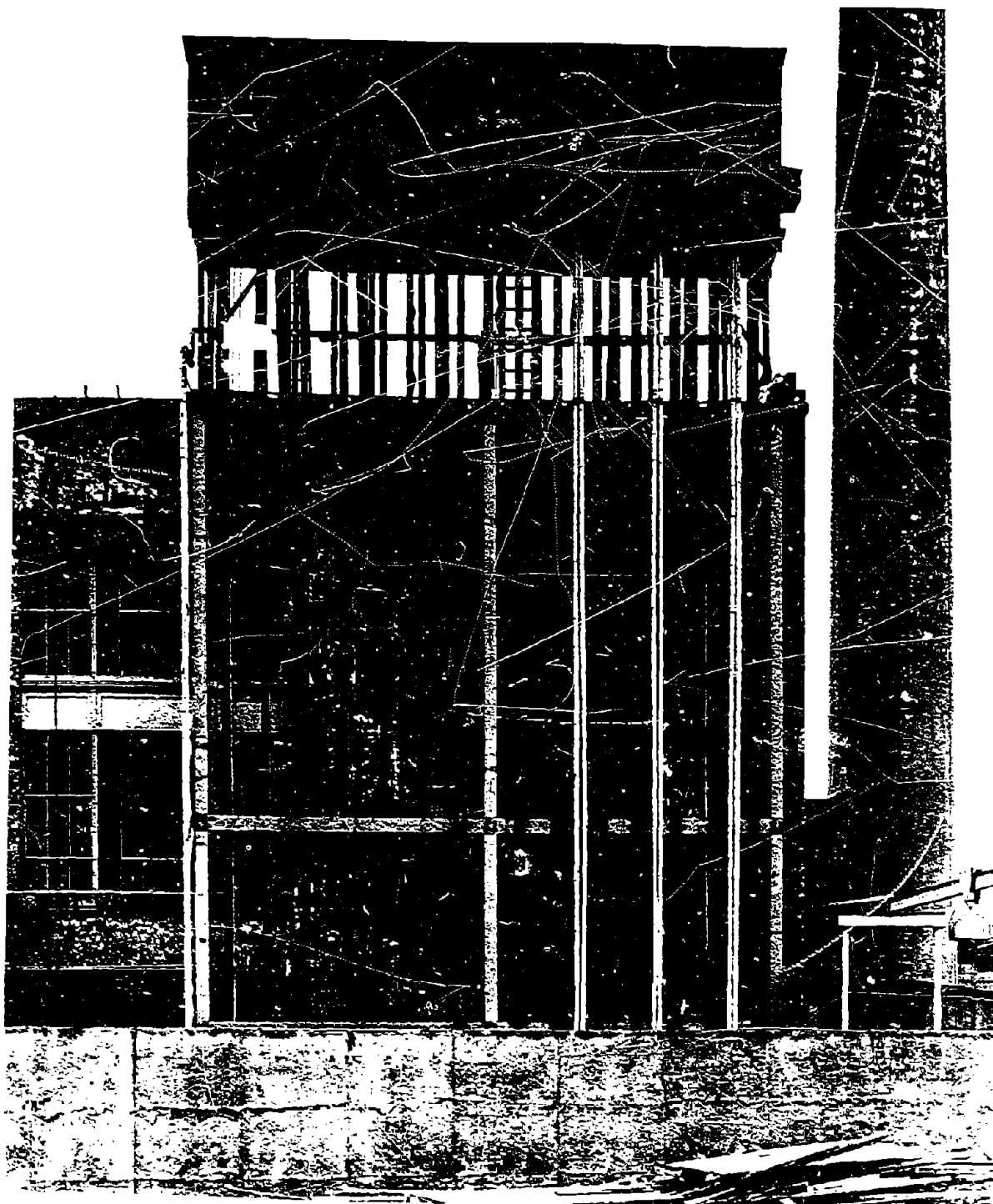
<u>Temperature</u>	<u>Density</u>
77°C	3.63
156°C	3.11
203°C	2.62
215°C	2.32
225°C	2.09
230°C	1.63

Some experiments have been run at temperatures of 275°C and densities of 2.1 gms/cm<sup>3</sup> in which the  $\text{UF}_6$  was in the gaseous state.

68. In passing it should be noted that, while a precise measurement of the change of density on melting has not been made, that this change amounts to roughly 20%. Uranium hexafluoride becomes more dense on freezing. A further constant that is of interest is viscosity of the liquid compound. This has been measured somewhat crudely and a result of 1.6 centipoises has been obtained for the liquid at 100°C.

69. An experiment was conducted to test electrical conductivity of  $\text{UF}_6$ . The material does not readily conduct either A.C. or D.C. and has a high electrical resistance (greater than  $10^9$  ohm cm<sup>s</sup>.)

SECRET



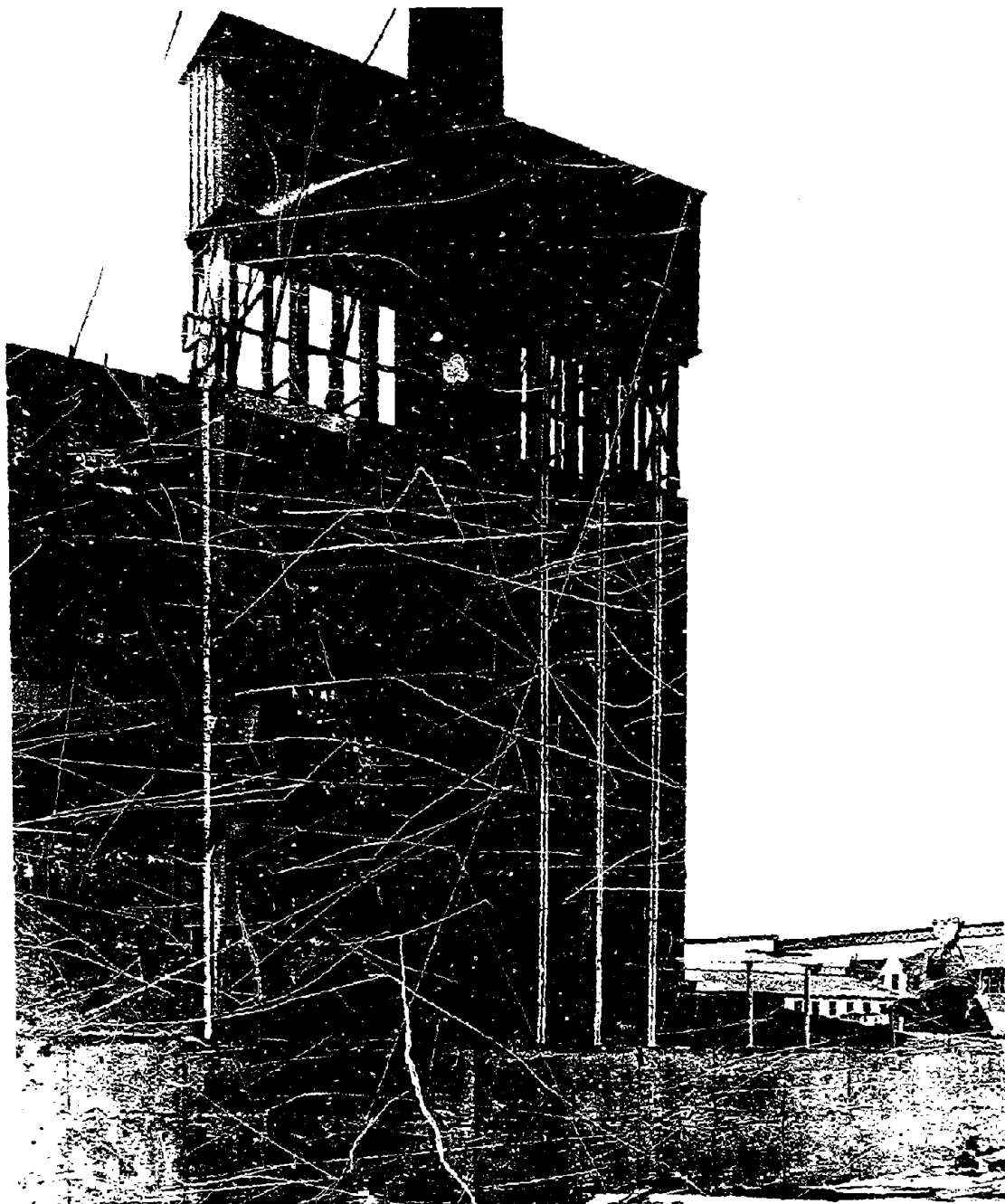
VIEW SHOWING UPPER PORTION OF PILOT PLANT

SECRET

View Showing Upper Portion of Pilot Plant

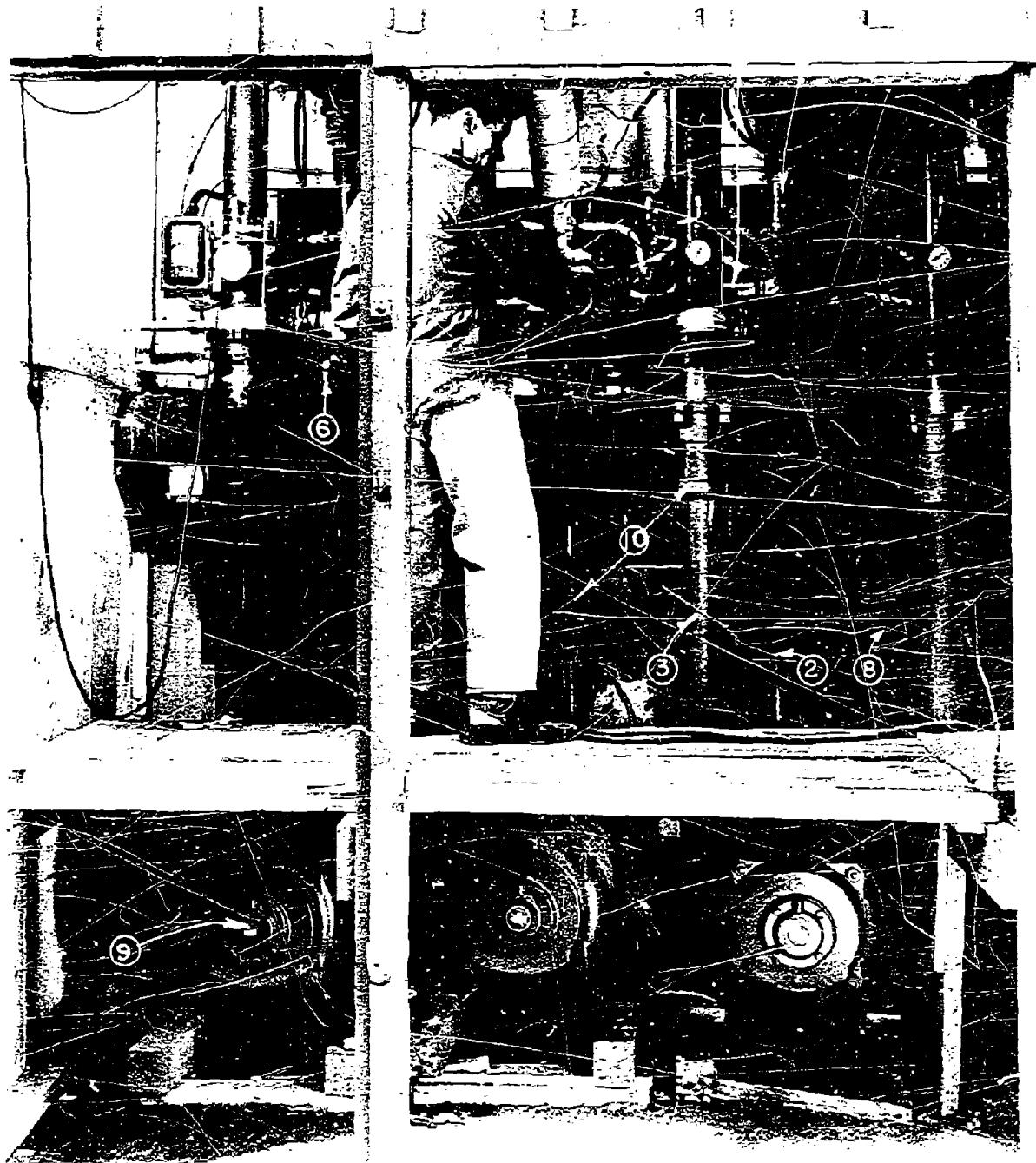
The lower ten feet is screened by a wall. Eight columns are in place on the front side of the structural steel. The three pairs of lagged lines on the right side of the picture carry 1/16" I.D. tubing which serves to connect columns in series. The 4" I.P.S. pipe behind the structural steel serves as the return portion of the circulating water loop.

SECRET



VIEW SHOWING UPPER PORTION OF PILOT PLANT





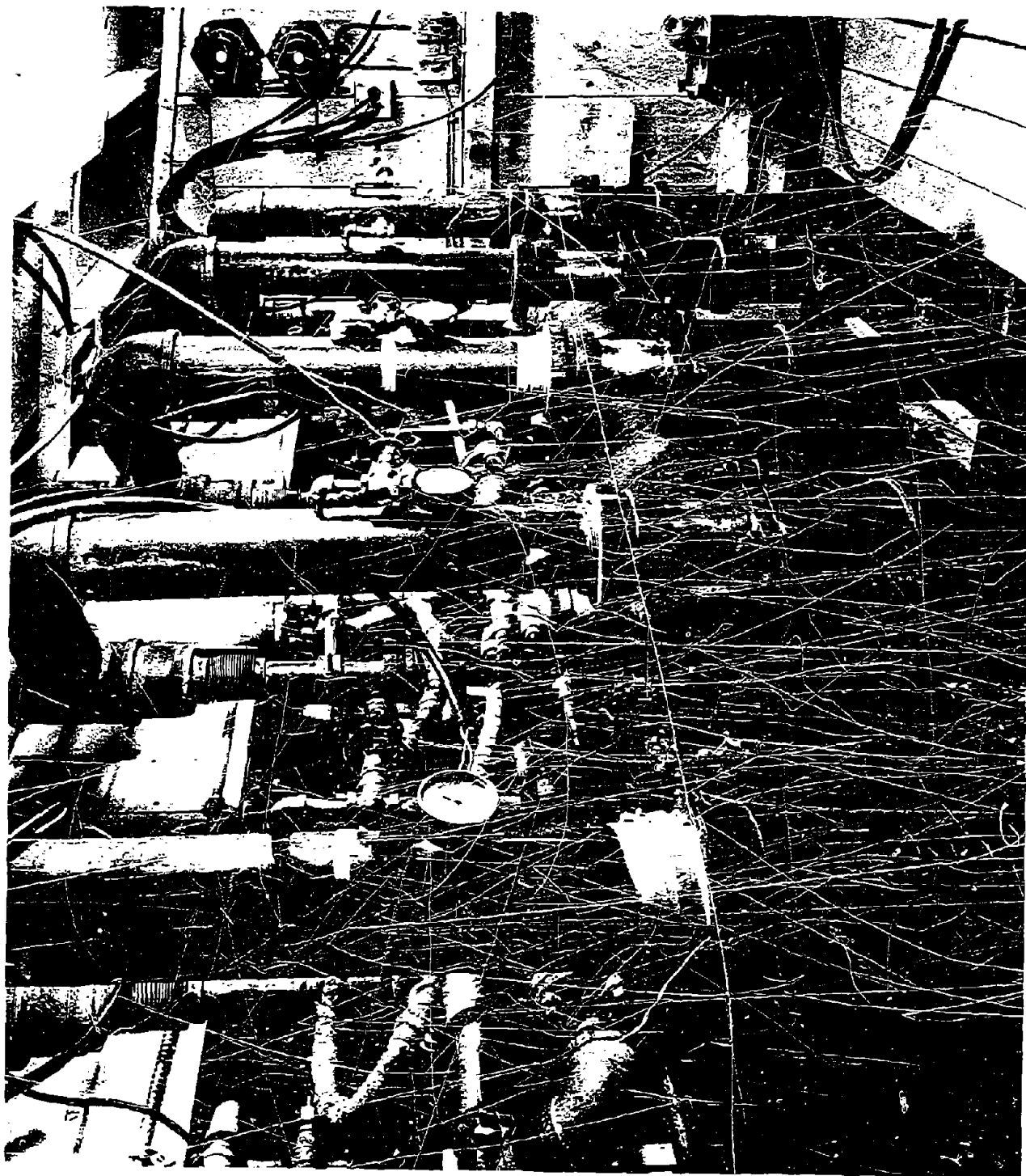
VIEW SHOWING BASE OF PILOT PLANT

SECRET

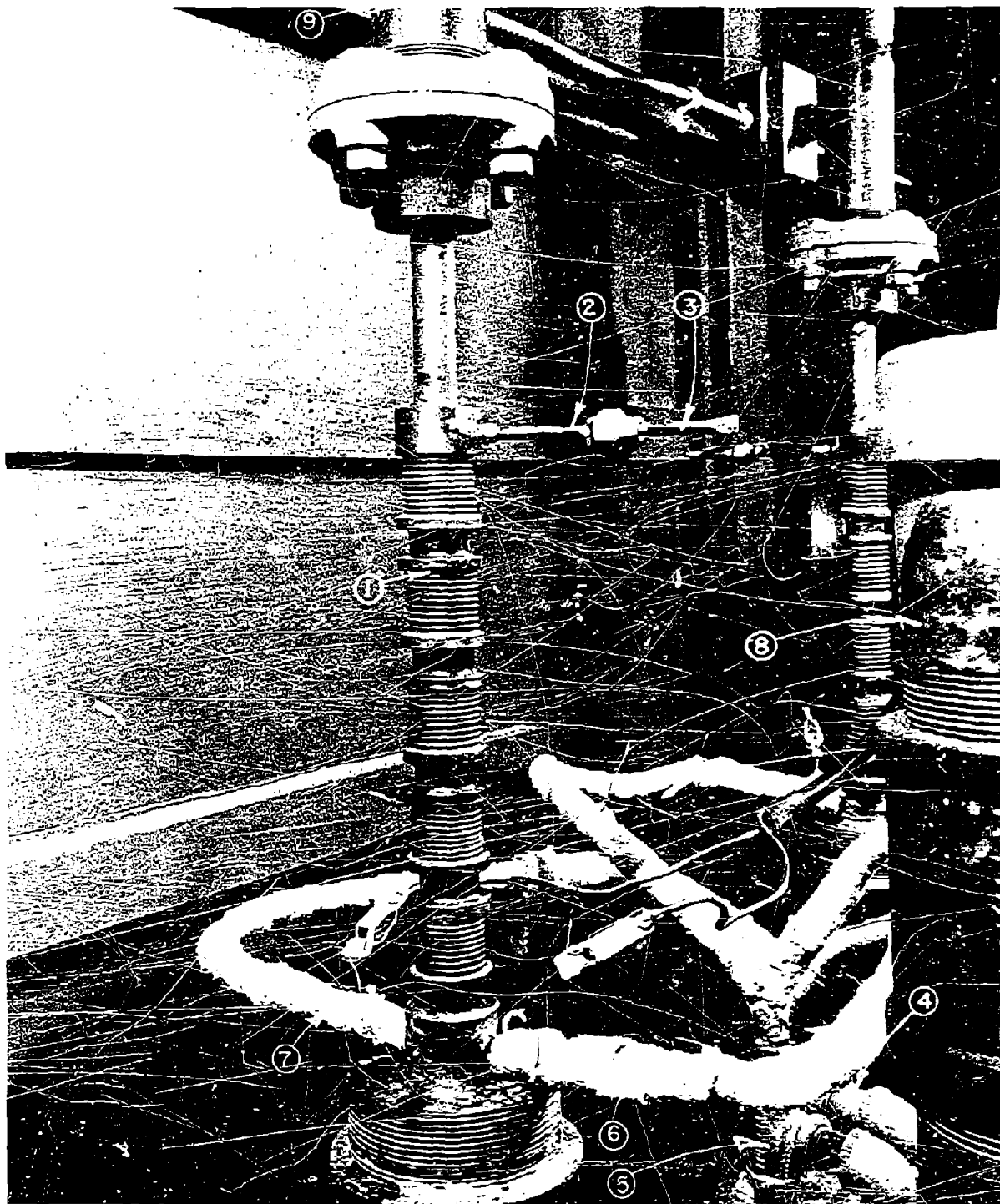
View Showing Base of Pilot Plant

1. Scientist Removing Sample of  $UF_6$
2. Circulating Water Returns
3. Circulating water input
4. Base of column
5. Steam line showing joint at base of column
6. Steam shut-off Valve
7. Steam Condensate Manifold
8. Water input Manifold
9. Centrifugal water circulating pumps
10. Joints allowing for thermal expansion
11. Storage Chamber for  $UF_6$

SECRET



VIEW AT BOTTOM OF PILOT PLANT



VIEW AT TOP OF COLUMN

SECRET

View at Top of Column

1. Sylphon bellows assembly which permits motion of hot nickel tubing relative to cold copper tubing.
2. Outlet tube for samples
3. Small Sample Container
4. Portion of connective loop connecting top of column at left to bottom of next column.
5. Pipe housing hot side of connective loop. This pipe carries two 1/16" I.D. tubes.
6. Pipe housing cold side of connective loop. This carries two 1/16" I.D. tubes.
7. Link Connecting column to hot side of loop.
8. Link Connecting column to cold side of loop. Electrically heated.
9. Top of circulating water return line
10. Steam input line.

SECRET

NOTE: ALL RIGHTS RESERVED THIS  
PAGE BEING PROTECTED

SILVER SOLDER IN PLACE  
AFTER CENTERING ANODE TUBE

NOTE: DIMENSIONS A  
3 - 45-16725

SWITCH SOLID IN PLACE  
AFTER CENTERING INNER TUBE

## LOWER SECTION

Copy available to DSC does not  
include the above information

# ASFRAY

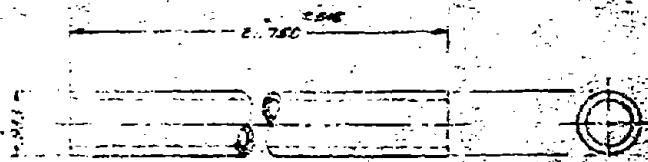


NOTE: DIMENSIONS A PLUS  
B = 35-10.125" IN



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NRLM



- ① TUBE  
NICKEL TUBING, 1000 O.D.  
SYMBOL 22  
1 REQ'D

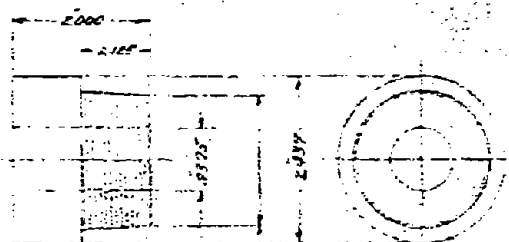
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SCALE 1/4" = 1 FT.



- ② SYLPHON RING  
NICKEL  
SYMBOL 3  
1 REQ'D

SLIDE FIT ON FT. 1



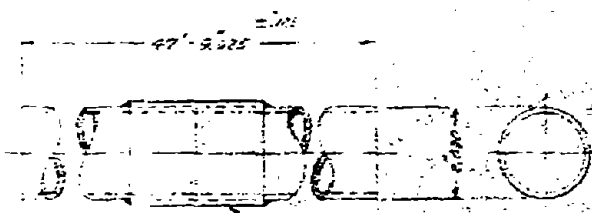
- ③ BUFFER COUPLING  
C.R. STEEL  
SYMBOL 52  
1 REQ'D

2 IPS. THREAD  
SLIDE FIT ON FT. 1

SCALE 1/4" = 1 FT.



- ④ SLEEVE  
NICKEL  
SYMBOL 53  
6 REQ'D



- ⑤ OUTER TUBE  
NICKEL TUBING, HARD DRAWN  
SYMBOL 23  
1 REQ'D

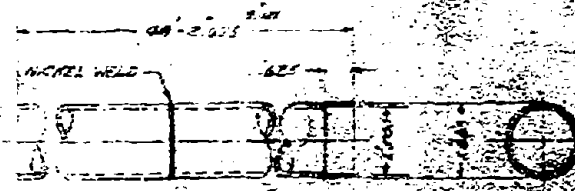
WHEN SPLICING TUBE TO BRASS REQ'D. LENGTH, USE COPPER SLEEVES ON OUTSIDE OF TUBE THERE TO BE 6 INCHES AND 2.000 O.D. SOLDER SLEEVE & TUBE TOGETHER

SCALE 1/4" = 1 FT.



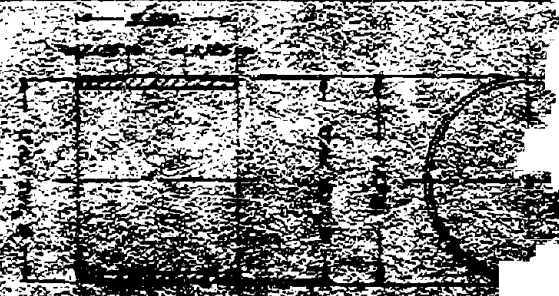
- ⑥ SPACER  
NICKEL  
SYMBOL 54  
1 REQ'D

SLIDE FIT ON FT. 1



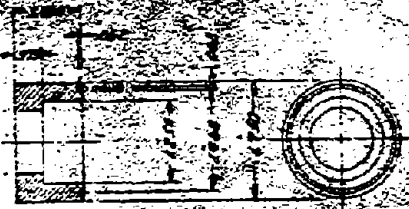
- ⑦ INNER TUBE  
NICKEL TUBING, 1000 O.D.  
SYMBOL 22  
1 REQ'D

WHEN SPLICING TUBE TO BRASS REQ'D. LENGTH, USE COPPER SLEEVES ON OUTSIDE OF TUBE THERE TO BE 6 INCHES AND 2.000 O.D. SOLDER SLEEVE & TUBE TOGETHER





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SLIDE FIT ON P.T. 1

3 SYLPHON MTG. RING  
NICEEL  
SYMBOL 3  
1 REGRD

SCALE 9/16 INCH

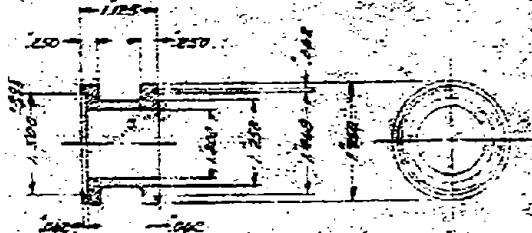


SLIDE FIT ON P.T. 1

SLIDE FIT ON P.T. 6

5 CONNECTOR  
COPPER BRASS  
SYMBOL 5  
1 REGRD

SCALE 9/16 INCH



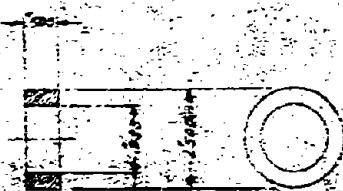
4 SLEEVE  
NICEEL  
SYMBOL 4  
6 REGRD

SCALE 9/16 INCH



6 TEE  
NICEEL  
SYMBOL 6  
1 REGRD

SCALE 9/16 INCH



9 SPACER  
NICEEL  
SYMBOL 9  
1 REGRD

SCALE 9/16 INCH



7 COUPLING  
NICEEL  
SYMBOL 7  
1 REGRD

SCALE 9/16 INCH

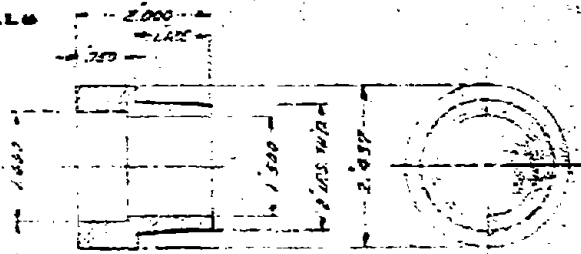
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permit fully legible reproduction



HEAT EXCHANGER

PLATE

REL 6



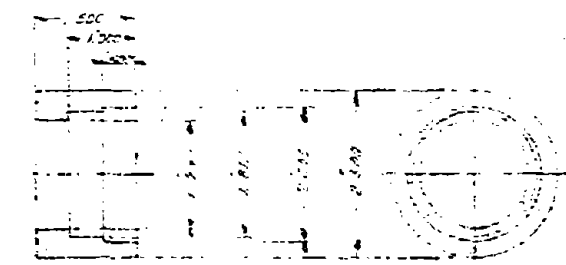
16 LOWER COUPLING  
C.B. STEEL  
SYMBOL 33  
1 REQD

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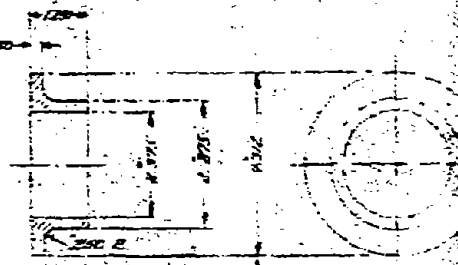
SCALE 2:1 (1/2")



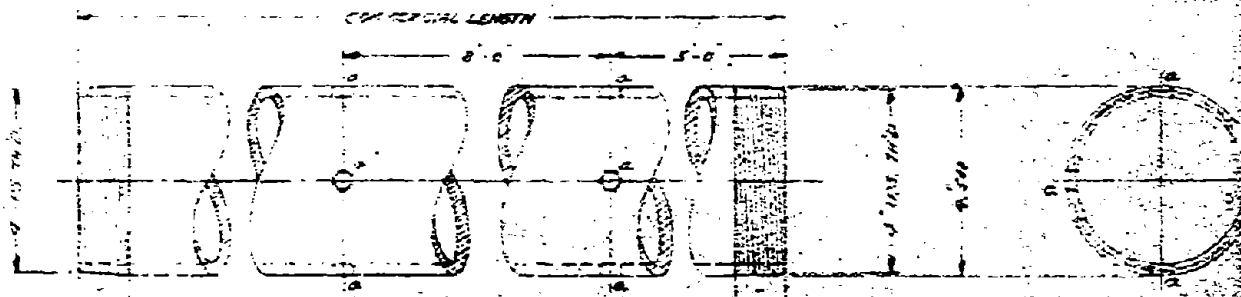
18 SYLPHON MTE. RING  
COPPER, HARD DRAWN  
SYMBOL 34  
1 REQD



17 7/8" MOUNTING  
COPPER, HARD DRAWN  
SYMBOL 35  
1 REQD



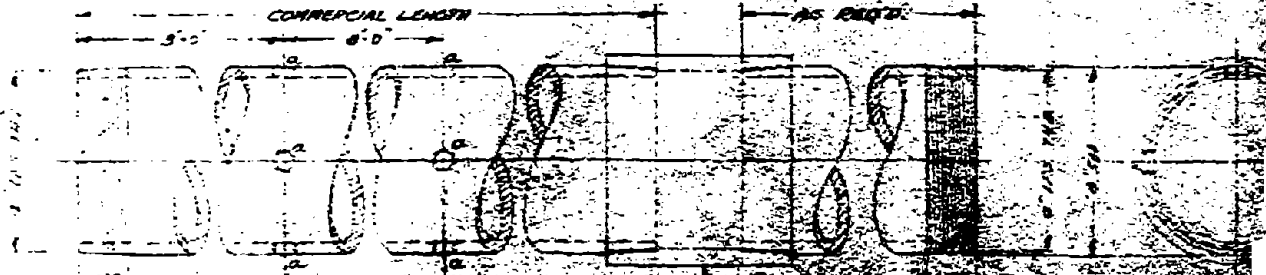
19 MTE. FLANGE  
BERS (CONSOLE-1)  
SYMBOL 36  
1 REQD



22 HOUSING  
4" STANDARD STEEL PIPE  
SYMBOL 38  
1 REQD

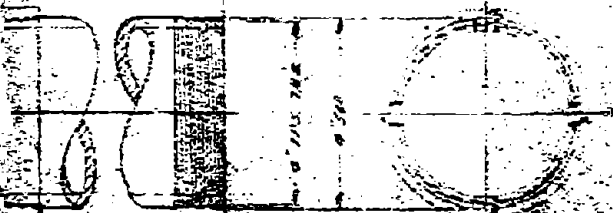
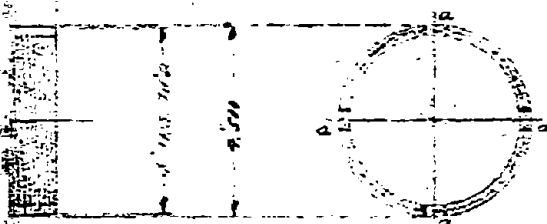
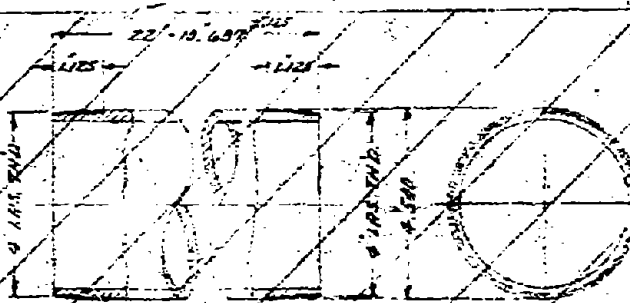
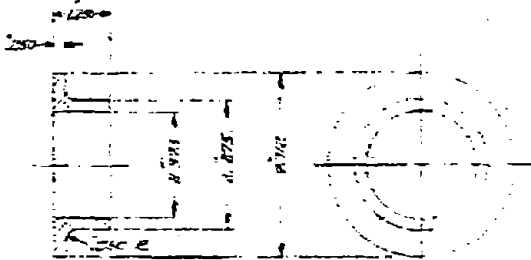
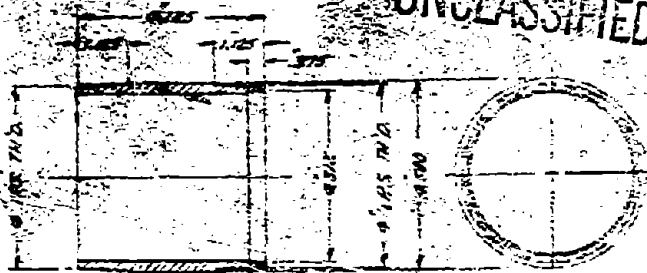
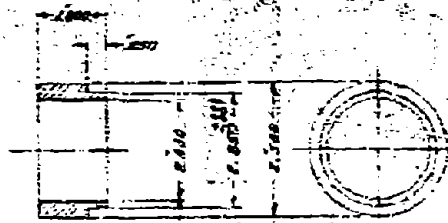
Q-2-20 7/8" - 6 INCHES

NOTE: - TOTAL LENGTH OF FT 22  
PLUS FT 23 TO BE 48 FT 9.375 IN.



23 HOUSING  
4" STANDARD STEEL PIPE  
SYMBOL 39  
1 REQD

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## HEAT EXCHANGER

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PLATE 8